Wigner crystallization of photons in cold Rydberg ensembles

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The coupling of weak light fields to Rydberg states of atoms under conditions of electromagnetically induced transparency (EIT) leads to the formation of Rydberg polaritons which are quasi-particles with tunable effective mass and long-range interactions. Confined to one spatial dimension their low energy physics is that of a moving-frame Luttinger liquid which due to the long-range character of the repulsive interaction can form a Wigner crystal. We calculate the Luttinger K parameter using density-matrix renormalization group (DMRG) simulations and find that under typical slow-light conditions kinetic energy contributions are too strong for crystal formation. However, adiabatically increasing the polariton mass by turning a light pulse into stationary spin excitations allows to generate true crystalline order over a finite length. The dynamics of this process and asymptotic correlations are analyzed in terms of a time-dependent Luttinger theory.

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The extraordinary properties of Rydberg atoms [1], such as large dipole-dipole interactions and long life-times, are currently attracting much attention. The interest ranges from quantum information [2–4] to many-body phenomena [5–15]. So far only few works considered the effect of interactions onto the light fields [16–20]. In recent experiments [16] it was shown that under EIT conditions the Rydberg interaction leads to a non-local, and strongly non-linear behaviour of the probe field [22, 23]. This gives rise to, e.g., the formation of a small avoided volume which contains at most one excitation [20], which can be described by considering only two excitations [3]. In the present paper we want to explore the many-body properties on larger length scales. One of the simplest but most dramatic effects resulting from a long-range repulsive interaction is the formation of a Wigner crystal, predicted for electrons in the early days of quantum mechanics [25]. We will show that a similar phenomenon can be observed in a dilute 1D gas of photons coupled to Rydberg atoms, which is not possible in e.g. Kerr-type point-interacting systems [26, 27]

Under conditions of EIT and small excitation densities, the coupling between photons and Rydberg atoms leads to the formation of dark-state polaritons (DSP) [28, 29], which behave like massive particles with strong repulsive and long-range interaction. We analyze the formation of a quasi-crystalline state of polaritons in 1D using DMRG simulations and time-dependent Luttinger-liquid (LL) theory. We show that under slow-light conditions the moving-frame ground-state displays density-wave correlations which decay very fast in propagation direction due to the small polariton mass. However, converting them into stationary spin excitations by decelerating a light pulse to a full stop inside a gas of Rydberg atoms [30, 31] can lead to a state with perfect crystalline order over the length of the medium.

To be specific, we consider an ensemble of N atoms with a three-level linkage-pattern [cf. Fig. 1(b)], composed of a ground-state $|\mathrm{g}\rangle$, intermediate state $|\mathrm{e}\rangle$ and metastable Rydberg-state $|\mathrm{r}\rangle$. The transition $|\mathrm{g}\rangle - |\mathrm{e}\rangle$ is driven by a quantized probe field $\hat{E} = \sqrt{\frac{\hbar\omega_\mathrm{p}}{2\epsilon_0}}\hat{\mathcal{E}}(\mathbf{r},t)\mathrm{e}^{-i(\omega_\mathrm{p}t-\mathbf{q}_\mathrm{p}\mathbf{r})} + \mathrm{H.a.}$, with carrier frequency ω_p and wave-vector $\mathbf{q}_\mathrm{p}$. $\hat{\mathcal{E}}(\hat{\mathcal{E}}^\dagger)$ are normalized field amplitudes corresponding to annihilation (creation) of a photon and are slowly varying in space and time. The transition $|\mathrm{e}\rangle - |\mathrm{r}\rangle$ is coupled via a strong external control field with Rabi frequency Ω, carrier frequency ω_c and wave-vector $\mathbf{q}_\mathrm{c}$. We chose the z-axis as the common propagation direction of the probe and control field and define the one- and two-photon detunings as $\Delta = \omega_\mathrm{e} - \omega_\mathrm{g} - \omega_\mathrm{p}$, $\delta = \omega_\mathrm{r} - \omega_\mathrm{g} - \omega_\mathrm{c} - \omega_\mathrm{p}$, respectively, where $\omega_{\mathrm{g,e,r}}$ are the energies of the corresponding atomic states ($\hbar = 1$).

In the absence of Rydberg interactions the system Hamiltonian can be diagonalized using adiabatic eigen-solutions, the dark- and bright-state polaritons (BSP), which fulfill approximate bosonic commutation relations [28, 29]. Following [32] we define the DSPs as $\hat{\Psi} = \cos\theta\hat{\mathcal{E}} - \sin\theta\hat{\Sigma}_\mathrm{gr}\mathrm{e}^{i(q_\mathrm{c}+q_\mathrm{p})z}$, and BSPs as $\hat{\Phi} = \sin\theta\hat{\mathcal{E}} + \cos\theta\hat{\Sigma}_\mathrm{gr}\mathrm{e}^{i(q_\mathrm{c}+q_\mathrm{p})z}$ where $\tan^2\theta = g^2 n/\Omega^2$. Here $\hat{\Sigma}_{\mu\nu}$ are continuous atomic spin flip operators, and n is the atom density. The DSP propagates loss-

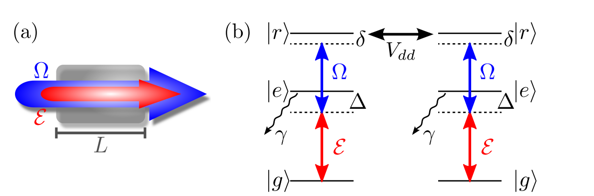

Figure 1. (a) Schematic setup for the creation of dark-state polaritons in a medium on length L. (b) Effective atomic linkage pattern for EIT in Rydberg gases. The weak quantized field $\hat{\mathcal{E}}$ is off-resonantly driving the $|\mathrm{g}\rangle - |\mathrm{e}\rangle$ transition with a one-photon detuning Δ, whereas the strong control field Ω is driving the $|\mathrm{e}\rangle - |\mathrm{r}\rangle$ transition with a final two-photon detuning δ.

less with group velocity $v_{\rm g} = c\cos^2\theta$, while the BSP has a velocity $c\sin^2\theta$ and is subject to losses with rate Ω_e^2/Γ, where $\Gamma = \gamma + i\Delta$, with 2γ being the spontaneous decay rate of state $|{\rm e}\rangle$, and $\Omega_e^2 = g^2 n + \Omega^2$. Near single-photon resonance, i.e., $|\Delta| \le \gamma$, and for an optically thick medium, i.e., $L \gg L_{\rm abs}$, where $L_{\rm abs} = g^2 n/c\gamma$ is the resonant absorption length in absence of EIT, an input bright-polariton will quickly be damped out. In the following we will consider the opposite case $|\Delta| \gg \gamma$, where absorption is irrelevant. However if $\cos^2\theta \ll \sin^2\theta \approx 1$ and if light pulses of finite length are considered an input bright-polariton can be disregarded as it will quickly escape the medium. Thus the BSP can be eliminated and the free dynamics of the DSPs is governed after a short transient by [33]

$$\hat{H}_0 = \int \mathrm{d}^3\mathbf{r}\, \hat{\Psi}^\dagger(\mathbf{r}) \left[\frac{\hat{p}_z^2}{2m_\parallel} + \frac{\hat{\mathbf{p}}_\perp^2}{2m_\perp} - v_{\rm g}\hat{p}_z + \tilde{\delta}(\mathbf{r})\right] \hat{\Psi}(\mathbf{r}), \quad (1)$$

where $\hat{p}_z = -i\partial_z$, $\hat{\mathbf{p}}_\perp = -i\nabla_\perp$, and $\tilde{\delta}(\mathbf{r}) = \delta(\mathbf{r})\sin^2\theta$. This corresponds to an effective Schrödinger equation for particles with tensorial mass and additional drift term, moving in an external potential $\delta(\mathbf{r})\sin^2\theta$. The drift is determined by the EIT group-velocity $v_{\rm g}$, and the masses are $m_\parallel^{-1} = v_{\rm g} L_{\rm abs}\frac{\Delta}{\gamma}$ and $m_\perp^{-1} = \frac{v_{\rm g}}{2q_{\rm p}}$ [1, 32]. The above model is valid as long as the BSP amplitude is negligible and [33]

$$|\tilde{\delta}| \ll \frac{(\Omega^2 + g^2 n)}{|\Delta|}, \qquad \frac{L_{\rm abs}}{L_{\rm char}}\sin^4\theta \le \frac{\gamma}{|\Delta|} \quad (2)$$

The first condition describes the regime of perturbative coupling between DSP and BSP, the second denotes the region of slow-light dispersion [1, 29]. $L_{\rm char}$ is a characteristic length scale of the DSP.

Let us now take into account interactions between the atoms in their Rydberg-state $|{\rm r}\rangle$, $\hat{H}_{\rm Ryd} = \frac{1}{2}\sum_{j,j'} \hat{\sigma}_{\rm rr}^{(j)} V_{\rm dd}(\mathbf{r}_j - \mathbf{r}'_j)\hat{\sigma}_{\rm rr}^{(j')}$, where $V_{\rm dd}(\mathbf{r}) = C_\alpha/|\mathbf{r}|^\alpha$ with $\alpha = 6$ ($\alpha = 3$) being the van-der-Waals (resonant dipole) interaction. In the continuum limit and transforming to polaritons, we find to lowest order in $\cos\theta$

$$\hat{H}_{\rm int} = \frac{\sin^4\theta C_\alpha}{2} \int \mathrm{d}^3\mathbf{r}\mathrm{d}^3\mathbf{r}'\, \frac{\hat{\Psi}^\dagger(\mathbf{r})\hat{\Psi}^\dagger(\mathbf{r}')\hat{\Psi}(\mathbf{r}')\hat{\Psi}(\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|^\alpha}. \quad (3)$$

The effect of the interaction is equivalent to a two-photon detuning. Consequently, the interaction shift has to be smaller than $\Omega_e^2/|\Delta|$ which can be translated into a minimal distance $a_{\rm c} = (C_\alpha\gamma/\Omega_e^2)^{1/\alpha}$ the DSPs have to keep to ensure the validity of the model. As shown in [3] for the case of a resonant interaction (i.e., $\Delta = 0$) and large optical depth, an incoming coherent light pulse will quickly develop strong anti-bunching with a minimum separation length in propagation direction corresponding to the EIT blockade radius $a_{\rm b} = (C_\alpha\gamma/\Omega^2)^{1/\alpha}$. A similar effect happens for $\Delta \neq 0$ due to the fast escape of the BSP. Since under slow-light conditions $\cos\theta = \Omega/\Omega_e \ll 1$ the initial preparation produces DSPs with a mutual distance larger than the critical value $a_{\rm c}$ and a vacuum of BSPs. Consequently the system can be well described by $\hat{H} = \hat{H}_0 + \hat{H}_{\rm int}$.

To address the question whether the interaction leads to Wigner-crystallization of polaritons we restrict ourselves to one dimension (1D). This can be achieved e.g., by using elongated cigar-shaped atomic ensemble with transverse extent smaller than the blockade radius [20], or atoms in hollow-core fibers [35, 36] or trapped in the evanescent field of ultra-thin optical fibers [37, 38]. The low-energy physics can be described in terms of a Luttinger liquid (LL) [39]. The LL model allows for an exact treatment also in the case of bosons [40] with $1/|x|^\alpha$-interactions, as long as $\alpha > 1$. Transforming to a frame co-moving with the EIT group-velocity removes the drift term, $\propto v_{\rm g}\partial_z$, in eq. (1). Assuming a fixed number of excitations and working off single-photon resonance $|\Delta| \gg \gamma$, we follow the standard LL-approach [39] to construct an effective low-energy Hamiltonian

$$H_{\rm LL} = \frac{1}{2\pi}\int \mathrm{d}x \left\{ uK\left(\pi\hat{\Pi}\right)^2 + \frac{u}{K}\left(\nabla\hat{\phi}\right)^2 \right\}. \quad (4)$$

$\hat{\Pi}$ and $\hat{\phi}$ are conjugate fields with $\left[\hat{\phi}(x), \hat{\Pi}(y)\right] = i\delta(x-y)$. u and K are the sound velocity and the Luttinger parameter, respectively. The K-parameter governs the long-range behavior of the charge-density-wave correlations (CDW) in the ground-state. E.g., the oscillatory part of the density correlations is given by $\langle\hat{\rho}(z)\hat{\rho}(0)\rangle_{\rm osc} \sim \rho_0^2\cos(2\pi\rho_0 z)\, z^{-2K}$. Here $\hat{\rho}(z) = \hat{\Psi}^\dagger(z)\hat{\Psi}(z)$ and ρ_0 is the average density. As first-order correlations decay as $\langle\hat{\Psi}^\dagger(z)\hat{\Psi}(0)\rangle \sim z^{-1/(2K)}$ the point $K = 1/2$ marks the crossover from a regime where superfluid order dominates ($K > 1/2$) to a regime with predominant CDW correlations of period $1/\rho_0$ ($K < 1/2$).

At this point it should be noted that one can create true crystalline order by adding a weak periodic lattice potential $\delta(x) = \delta_0\sin(2\pi x/d)$, which leads to a sine-Gordon Hamiltonian [39] for commensurate fillings $\rho_0 = 1/(sd)$, $s \in \mathbb{N}$. This model exhibits a quantum phase transition to a gapped ordered phase for arbitrarily small but finite δ_0, if $K < K_s = 2/s^2$ [39, 41]. To avoid the necessity of a co-moving lattice potential one then should consider stationary-light polaritons [1, 42, 43].

Although no exact expression for K exists, an approximate closed formula was given in [44]:

$$K = \frac{1}{\sqrt{1+\eta_\alpha\Theta}}, \qquad \eta_\alpha = \frac{\alpha(\alpha+1)\zeta(\alpha)}{2\pi^2}. \quad (5)$$

Here $\zeta(x)$ is the Riemann Zeta-function and we introduced the dimensionless interaction strength $\Theta = \rho_0^{\alpha-2}\frac{mC_\alpha}{2\pi}$.

To check this expression we determined K numerically using DMRG [45] and made use of the fact that $K/u = \pi\rho_0^2\chi$ is determined by the compressibility $\chi^{-1} = \rho_0^2\frac{\partial\mu}{\partial\rho_0} = \rho_0^2 L\frac{\partial^2 E}{\partial N^2}$ [39]. Furthermore $uK = \pi\rho_0/m$, which is true for any Galilean-invariant model [46]. We have validated the numerical procedure for the case of the integrable Lieb-Liniger model, where χ can be calculated exactly as a function of interaction strength. Using a proper discretization of the model [47] leads to the results for K shown in Fig. 2. One recognizes

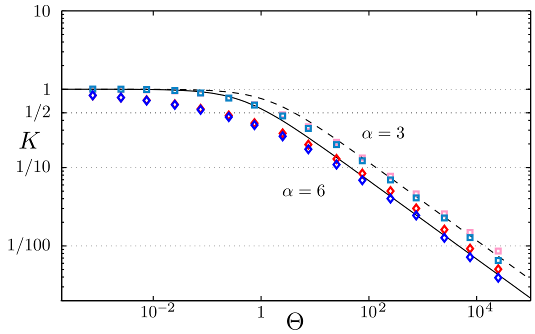

Figure 2. Luttinger parameter K as a function of interaction strength. The continuous (dashed) line shows the analytical approximation [44] for $\alpha = 6$ ($\alpha = 3$). Diamonds (squares) show results from DMRG calculations with open {red} and periodic {blue} boundary conditions (BC). The numerical parameters were $\Delta x = 1/10\rho_0$ with (i) $L = 20/\rho_0$ and bond dimension d=16 for open BC or (ii) $L = 10/\rho_0$ and d=32 for periodic BC. Quantum Monte Carlo results for $\alpha = 3$ can be found in [40].

good agreement for dipolar interactions ($\alpha = 3$) but a shift towards smaller values of K for vdW interactions ($\alpha = 6$). Nevertheless we will use the analytic expression in the following.

Let us first discuss the time-independent case $\Omega(t) = \Omega =$ const. We concentrate on the ground-state where CDW correlations should be most pronounced. In Fig. 3 we have plotted the normalized two-particle correlation $g^{(2)}(z)$ in the ground state of $\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}$ obtained by DMRG corresponding to different values of K. The large-z behavior is well described by LL correlations and one recognizes well pronounced oscillations for $K \leq 1/2$. For small distances the plots show an extended spatial region over which $g^{(2)}(z)$ vanishes. The correlations around $z = 0$ become more suppressed than in the case of free fermions, which is the strongest possible for point interactions [26, 48]. Thus the long-range interaction, eq. (3), prevents that two DSPs approach each other which also proves the consistency of the effective model.

We can use eq. (5) to estimate the critical interaction strength required to enter the CDW dominated regime, i.e., $K \leq 1/2$, giving $\eta_\alpha \Theta_{\text{crit}} = 3$. Θ is proportional to the effective mass of the polaritons $m \sim v_{\text{g}}^{-1} \sim \frac{g^2 n}{\Omega^2}$ which is different along longitudinal ($m_\parallel$) and transverse directions ($m_\perp$), and can be tuned via the control field Ω. Considering vdW interactions (and setting $\eta_6 = 1$) we find for $m = m_\parallel$.

$$\Theta_{\text{crit}} = \frac{1}{4\pi} \frac{\gamma}{|\Delta|} (\rho_0 L_{\text{abs}})^4 \, \text{OD}_B^6, \tag{6}$$

where we have introduced the optical depth per (initial) blockade radius $\text{OD}_B = a_{\text{b}}/L_{\text{abs}}$. Note that condition (2) translates into $\rho_0 L_{\text{abs}} \leq \gamma/|\Delta|$. Using e.g. $\rho_0 L_{\text{abs}} = \gamma/|\Delta| = 1/100$, we find that the optical depth per blockade at Θ_{crit} has to be $\text{OD}_B^\parallel \gtrsim 30$. This shows that a crystalline structure will be challenging to prepare along the propagation direction. Along the transvere direction the conditions are more relaxed. To overcome this difficulty we will now discuss an alternative based on light storage.

Let us consider an initial polariton pulse close to the ground state in the co-moving frame and time-dependent control fields. Decelerating the DSPs by reducing v_{g} in time preserves their spatial structure (and density ρ_0), in the absence of interactions [29]. At the same time their effective mass is increased, which suggests a decreasing K according to eq.(5) for interacting DSPs. When the pulse is brought to a complete stop, $K(t)$ approaches zero potentially leading to true long range order. If v_{g} is switched to zero instantaneously, the initial spatial correlations will be frozen. Thus the switching has to be done smoothly on a time scale τ long enough for correlations to propagate through the system. The latter process is determined by the speed of sound $u(t) = \pi\rho_0/(m(t)K(t))$. For small K we find the scaling $K \sim 1/\sqrt{m} \sim \sqrt{v_{\text{g}}}$, i.e., the speed of sound decreases only with the square root of the group velocity, $u(t) \sim 1/\sqrt{m(t)} \sim \sqrt{v_{\text{g}}}$.

In order to describe the adiabatic switch-off we consider the LL Hamiltonian (4) with time-dependent parameters $K(t)$ and $u(t)$ [49]. Using the standard decomposition of the fields into bosonic momentum modes $\hat{b}_p, \hat{b}_p^\dagger$ [39] yields

$$\hat{H} = \frac{1}{2} \frac{\pi\rho_0}{m(t)} \sum_{p\neq 0} |p| \Big\{ w(t) b_p^\dagger b_p - \frac{g(t)}{2} \Big(b_p^\dagger b_{-p}^\dagger + b_{-p} b_p \Big) \Big\}, \tag{7}$$

where $w(t) = 1 + 1/K(t)^2$, $g(t) = 1 - 1/K(t)^2$ and $K(t) = (1 + \eta_6 C_6 \rho_0^4 m(t)/2\pi)^{-1/2}$, according to eq. (5). The corresponding Heisenberg equations of motion can be diagonalized by a time-dependent Bogoliubov transformation. Since the transformation matrix is itself time-dependent this leads in general to an infinite hierarchy. However, if $K(t)$ and $u(t)$ are chosen such that $\dot{K}(t) \propto K(t)\, u(t)$ the hierarchy truncates after the second iteration. This requires $m(t) = m_0 f(t) = m_0 \, \mathrm{e}^{x(t)} \sinh[x(t)] \, \mathrm{e}^{-\text{arcosh}(C)}/\sqrt{C^2-1}$, with $x(t) = \text{arcosh}(t/\tau + C)$, where $C = (K_0^2 + 1)/(2K_0)$, $K_0 = K(t=0)$ and τ being the characteristic deceleration

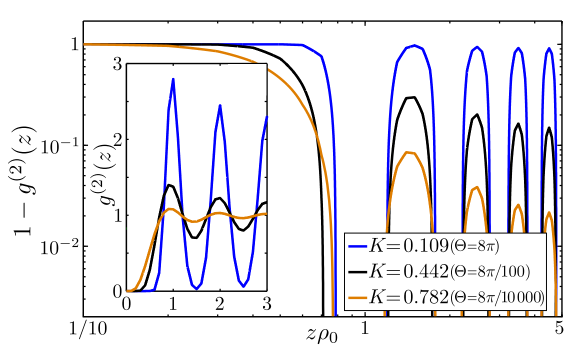

Figure 3. Normalized two-particle correlation. *main panel:* $1 - g^{(2)}(z)$, in double logarithmic scale. *inset:* $g^{(2)}(z)$ in linear scale. Full lines show numerical results for interaction strength increasing from orange over black to blue. The particles are subject to periodic boundary conditions. Note that at $\rho_0 z \gtrsim 5$ finite size effects become noticable.

time. The time-dependence of the control-field can then be computed as $\Omega(t) = g\sqrt{n}\big(f(t)\,c/v_g(0) - 1\big)^{-1/2}$, where $m_0 = m(t=0)$ and $m(t)^{-1} = v_g(t)L_{\rm abs}\Delta/\gamma$ was used. Then $K(t) = e^{-x(t)}$ and we find the oscillatory part of the equal time density-density correlations given by

$$\langle\hat\rho(z)\hat\rho(0)\rangle_{\rm osc} \sim \cos\big(2\pi\rho_0 z\big)\left(\frac{1}{\rho_0 z}\right)^{2K(t)} e^{-K(t)I(z,t)} \quad (8)$$

$$I(z,t) = \int_0^\infty \mathrm{d}p \frac{1-\cos pz}{p}\,\frac{\cos\xi(t) - 1 - \sqrt{l_0^2p^2-1}\sin\xi(t)}{1-l_0^2p^2},$$

where $\xi(t) = \sqrt{l_0^2p^2-1}\,\ln\big(K(t)/K_0\big)$. The first term $\sim z^{-2K(t)}$ describes the adiabatic behavior, where the spatial dependence of correlations is determined by the instantaneous value of $K(t)$. The exponential describes non-adiabatic corrections. As a consequence of the finite speed of sound, these corrections lead to a “crossover” from the power law with adiabatic exponent $K(t)$ to one with the initial exponent K_0 at the characteristic distance $l_0 = \eta_6 C_6 \rho_0^5\,\tau$, as can be seen from Fig. 4. Maximizing the switch-off time τ results in large spatial regions l_0 with quasi-crystalline order. On the other hand, τ has to be sufficiently small to bring the pulse to a complete stop after a length $L_d = \int_0^\infty dt v_{\rm g}(t)$ less than the medium length L. Setting $L_d = L$ and using the above protocol, we find

$$\frac{l_0}{L} = \frac{2\pi}{K_0}\rho_0 L_{\rm abs}\frac{|\Delta|}{\gamma} \quad (9)$$

Noting that K_0 is close to unity and that $\rho_0 L_{\rm abs} \le \gamma/|\Delta|$, this fraction can approach unity showing that a crystalline order over the whole medium is possible.

It should be noted that changing the control-field in time leads to an additional coupling between the DSP and BSP [29]. The decay rate due to this coupling is given by $\gamma_\theta = \gamma\dot\theta^2/g^2n$ and we need $\int_0^\tau \mathrm{d}t\gamma_\theta(t) \ll 1$. Using the above protocol we find $c\tau/L_{\rm abs} \gg 4K_0^2/(K_0^2-1)^2$. For $K_0 \approx 0.99$ and $L_{\rm abs} \approx 5\ \mu$m we have $\tau \gg 40\ \mu$s, which is certainly feasible.

So far we have assumed that the initial state for the light storage is the moving-frame groundstate of the Hamiltonian (7). Let us now discuss the effects of initial excitations. As the system is non-integrable it is reasonable to assume that the state of the DSPs after the initial preparation is thermal (we set $k_B = 1$). In a thermal state all correlations decay exponentially for distances larger than $L_T \sim u/T$, with correlation length $\xi_T \sim L_T/K = \pi\rho_0/(mTK^2)$ [50]. Since for a non-interacting gas and adiabatic mass changes $T(t) \sim 1/m(t)$ holds, one naïvely expects that the correlation length increases $\xi_T \sim 1/K^2(t)$ as long as one stays adiabatic. Evaluating the bosonic correlation functions in eq. (8) with a thermal distribution we find a slightly different result (cf. Fig. 4). Correlations decay as $\exp(-|z|^2/L_{\rm corr}^2)$ for length scales $|z| \lesssim L_{\rm corr} = 2\sqrt{l_0 L_T^0}/\pi K_0 \times (\ln(K_0/K(t)))^{1/4}$ which crosses over to an exponential decay for larger length scales. Here L_T^0 is the initial thermal correlation length.

To estimate the maximal initial temperature of the polariton system we observe that any polariton components with fre-

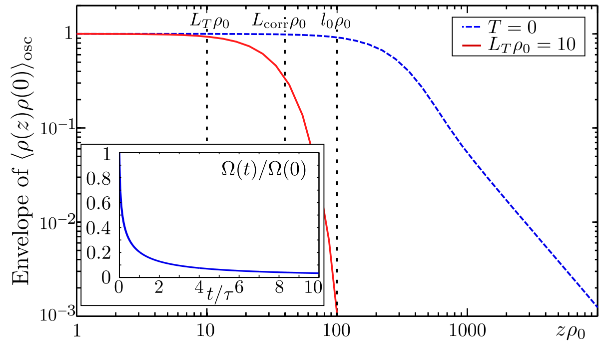

Figure 4. *main panel:* space dependent amplitude of the oscillatory part of the correlation function in the long time limit, $K_0 = 0.8$, $K(t) = 5\times10^{-5}$. The dashed blue line shows the spatial decay of density-density correlations for zero Temperature, which shows a crossover from adiabatic to diabatic algebraic decay at $l_0\rho_0 = 100$ (indicated by rightmost vertical line). The solid red line shows the modified decay for initial Temperature corresponding to a thermal length $L_T\rho_0 = 10$ (leftmost vertical line) which shows a crossover to exponential decay at length scale $L_{\rm corr}\rho_0 \approx 40$.
inset: $\Omega(t)/\Omega(0)$ for $K_0 = 0.8$ and $v_{\rm gr}(0)/c = 10^{-5}$.

quencies larger than the off-resonant EIT line-width $\Omega^2/|\Delta|$ will escape [33]. Thus we can give as a reasonable estimate for an upper limit $T \approx \frac{1}{2}\frac{\Omega^2}{|\Delta|}$. With this we find

$$\frac{\sqrt{l_0 L_T^0}}{L} = \left(\rho_0 L_{\rm abs}\frac{|\Delta|}{\gamma}\right)\sqrt{\frac{2\pi}{\rm OD}\frac{|\Delta|}{\gamma}} \quad (10)$$

which gives a measure for the final correlation length in units of the total length. Although the first term on the right side is less than unity, the whole expression can approach unity.

In summary we showed that the combination of EIT with interacting Rydberg gases leads to strongly interacting light-matter particles, termed Rydberg polaritons. We discussed the experimental requirements needed to obtain a quasi-long-range-ordered ground-state corresponding to a moving-frame Wigner crystal of Rydberg excitations in 1D by mapping the problem to a Luttinger liquid. Numerical and analytic results showed that under slow-light conditions the kinetic energy contributions in the longitudinal direction are too large to enter the density-wave dominated regime. Using a time-dependent Luttinger liquid approach we showed, however, that decelerating a light pulse in a gas of Rydberg atoms to a full stop over a sufficiently long deceleration time can create true crystalline order over a substantial fraction of the medium. Turning the Wigner crystal of spin excitations back into electromagnetic fields by a sudden switch-on of the drive field [28] produces a train of photons with long range crystalline order.

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WIGNER CRYSTALLIZATION OF PHOTONS IN COLD RYDBERG ENSEMBLE – SUPPLEMENTAL MATERIAL

In this supplementary we derive the effective Hamiltonian (1) of the main text in one spatial dimension and explicitly establish its conditions of validity. The treatment is based on the formalism developed in [1]. The interaction of probe and control fields with the three-level atoms shown in Fig. 1 of the main text in the absence of Rydberg interactions can be described by the following atom-light coupling Hamiltonian in a rotating frame

$$\hat{H} = \int \mathrm{d}^3\mathbf{r} \Big\{ \Delta\hat{\Sigma}_{ee}(\mathbf{r}) + \delta\hat{\Sigma}_{rr}(\mathbf{r}) + \Omega\hat{\Sigma}_{re}\mathrm{e}^{i\mathbf{q}_c\mathbf{r}} + g\sqrt{n}\hat{\mathcal{E}}(\mathbf{r})\hat{\Sigma}_{eg}(\mathbf{r})\mathrm{e}^{i\mathbf{q}_p\mathbf{r}} + \text{H.a.}\Big\}, \tag{11}$$

where all quantities are defined as in the main text and $\hat{\Sigma}_{\mu\nu}(\mathbf{r}) \equiv \sum_{j\in\Delta V} |\mu\rangle_{jj}\langle\nu|/\sqrt{\Delta N}$ are continuous atomic flip operators defined on a small volume $\Delta V(\mathbf{r})$ centered around position $\mathbf{r}$ containing $\Delta N \gg 1$ atoms. Assuming that all atoms are initially prepared in the ground state $|g\rangle$ and considering weak probe fields, i.e. a photon density much less than the atom density, we can treat the light-atom coupling perturbatively. Consequently, in lowest order of the atom-field coupling g we find the Heisenberg-Langevin equations for the atomic operators

$$\frac{\partial}{\partial t}\hat{\Sigma}_{ge} = -(i\Delta+\gamma)\hat{\Sigma}_{ge} + \hat{F}_{ge} + ig\sqrt{n}\,\hat{\mathcal{E}}\mathrm{e}^{i\mathbf{q}_p\mathbf{r}} + i\Omega^*\,\hat{\Sigma}_{gr}\mathrm{e}^{-i\mathbf{q}_c\mathbf{r}}, \tag{12}$$

$$\frac{\partial}{\partial t}\hat{\Sigma}_{gr} = -i\delta\hat{\Sigma}_{gr} + i\Omega\,\hat{\Sigma}_{ge}\mathrm{e}^{i\mathbf{q}_c\mathbf{r}}. \tag{13}$$

Here the $\hat{F}_{ge}$ is a delta-correlated Langevin noise operator associated with the decay from the intermediate (excited) state $|\mathrm{e}\rangle$ which is necessary to preserve commutation relations [2]. One easily verifies that the correlation functions of the Langevin operators are proportional to the population in the excited state $|e\rangle$. In the linear response and for sufficiently small two-photon detuning δ this population is small and we can safely ignore the noise operators in the following. If need be these operators can be re-introduced by hand using the fluctuation-dissipation theorem. To arrive at a closed description of the atom-field system we also need the equation of motion for the slowly varying probe-field envelope $\hat{\mathcal{E}}(\mathbf{r},t)$. Restricting ourselves to a one-dimensional problem the dynamics of the probe field is described by a truncated wave-equation in paraxial approximation

$$\left[\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right]\hat{\mathcal{E}}(z,t) = ig\sqrt{n}\,\hat{\Sigma}_{ge}(z,t)\mathrm{e}^{i\mathbf{q}_p\mathbf{r}}. \tag{14}$$

Transforming Eqs. (12-14) into Fourier space according to $f(z,t) = \int \mathrm{d}k\, e^{-ikz} f(k,t)$ yields the following matrix equations

$$\frac{d}{dt}\mathbf{X} = -i\mathsf{H}\mathbf{X} \tag{15}$$

where $\mathbf{X}^\top = \{\hat{\mathcal{E}}, \hat{\Sigma}_{gr}\mathrm{e}^{i(\mathbf{q}_p+\mathbf{q}_c)\mathbf{r}}, \hat{\Sigma}_{ge}\mathrm{e}^{i\mathbf{q}_p\mathbf{r}}\}$ and the Hamiltonian matrix reads

$$\mathsf{H} = \begin{bmatrix} -kc & 0 & -g\sqrt{n} \\ 0 & \delta & -\Omega \\ -g\sqrt{n} & -\Omega^* & \Delta - i\gamma \end{bmatrix}.$$

Changing the basis to a description in terms of dark- and bright-polaritons $\mathbf{Y}^\top = \{\hat{\Psi}, \hat{\Phi}, \hat{\Sigma}_{ge}\mathrm{e}^{i\mathbf{q}_p\mathbf{r}}\}$ via $\hat{\Psi} = \cos\theta\hat{\mathcal{E}} - \sin\theta\hat{\Sigma}_{gr}\mathrm{e}^{i(\mathbf{q}_p+\mathbf{q}_c)\mathbf{r}}$ and $\hat{\Phi} = \sin\theta\mathcal{E} + \cos\theta\hat{\Sigma}_{gr}\mathrm{e}^{i(\mathbf{q}_p+\mathbf{q}_c)\mathbf{r}}$ yields the equation of motion $\partial_t\mathbf{Y} = -i\mathsf{H}'\,\mathbf{Y}$ with

$$\mathsf{H}' = \begin{bmatrix} \delta\sin^2\theta - kc\cos^2\theta & -\sin\theta\cos\theta(\delta+kc) & 0 \\ -\sin\theta\cos\theta(\delta+kc) & \delta\cos^2\theta - kc\sin^2\theta & -\Omega_e \\ 0 & -\Omega_e & \Delta - i\gamma \end{bmatrix}.$$

Assuming that the time evolution is slow compared to $|\Delta - i\gamma|$ we can adiabatically eliminate the optical polarization $\hat{\Sigma}_{ge}$ which yields the coupled equations for bright and dark polaritons

$$\frac{d}{dt}\begin{bmatrix}\hat{\Psi}\\ \hat{\Phi}\end{bmatrix} = -i\mathsf{H}''\begin{bmatrix}\hat{\Psi}\\ \hat{\Phi}\end{bmatrix} \tag{16}$$

with

$$\mathsf{H}'' = \begin{bmatrix} \delta\sin^2\theta - kc\cos^2\theta & -\sin\theta\cos\theta(\delta+kc) \\ -\sin\theta\cos\theta(\delta+kc) & \delta\cos^2\theta - kc\sin^2\theta - \frac{\Omega_e^2}{\Delta - i\gamma} \end{bmatrix}.$$

For $\Delta > 0$ and under slow-light conditions, i.e. $\sin^2\theta \gg \cos^2\theta$, one recognizes that the off-diagonal coupling terms are small compared to the difference of the diagonal elements. Under these conditions the dynamics of dark and bright polaritons approximately separates and one can treat their cross coupling perturbatively. Within this perturbative treatment the effective equation of motion of the dark polariton $\hat{\Psi}$ up to second order of the off-diagonal coupling is given by

$$\frac{d}{dt}\hat{\Psi} = -i\left(\delta\sin^2\theta - kc\cos^2\theta\right)\hat{\Psi} - i\frac{\sin^2\theta\cos^2\theta(\delta+kc)^2}{(\delta+kc)(\sin^2\theta-\cos^2\theta) + \frac{\Omega_e^2}{\Delta - i\gamma}}\hat{\Psi}.$$

To arrive at an even more simplified but more transparent equation we assume $\delta \geq 0$ and require $\delta + kc \ll \Omega_e^2/|\Delta|$ for all relevant values of k, which implies in particular

$$0 \leq \delta \ll \frac{\Omega_e^2}{|\Delta|}, \tag{17}$$

where we used $|\Delta| \gg \gamma$. In this limit we find that the dynamics of the DSPs is described by

$$\frac{d}{dt}\hat{\Psi} = -i\delta\left(1 + \frac{\delta\Delta\cos^2\theta}{\Omega_e^2}\right)\hat{\Psi} + ikv_{\mathrm{g}}\left(1 - 2\frac{\delta\Delta}{\Omega_e^2}\right)\hat{\Psi} - i\frac{v_{\mathrm{g}}c\Delta}{\Omega_e^2}k^2\hat{\Psi}, \tag{18}$$

where we approximated $\sin^2\theta \approx 1$. The first term on the right hand side describes an energy offset due to a finite two-photon detuning. The second term accounts for the propagation with group velocity $v_{\rm g} = c\cos^2\theta$. The third term describes the quadratic dispersion with effective mass $m_{\|}^{-1} = c^2\cos^2\theta|\Delta|/\Omega_e^2 \approx v_{\rm g}L_{\rm abs}|\Delta|/\gamma$.

Condition (17) also determines the validity of the interaction Hamiltonian (3) of the main text. As already pointed out in the main text, the Rydberg interactions effectively induce a space-dependent two-photon detuning [3] which combined with eq. (17) leads to the critical minimal distance $a_{\rm c} = (C_\alpha\gamma/\Omega_e^2)^{1/\alpha}$.

We can also interpret the validity of the perturbation theory as a condition of a maximal k-value until which a separation into DSPs with slow-light dispersion and fast moving BSPs is valid. This condition reads

$$|kc| \ll \frac{\Omega_e^2}{|\Delta|} \tag{19}$$

leading to $|k_{\rm max}| = \Omega_e^2/|\Delta|c$. Plugging this into the zeroth order dispersion relation of the DSP leads to the maximal energy $\omega_{\rm max} = v_{\rm g}k_{\rm max} = \Omega^2/|\Delta|$, which in the end determines the maximal temperature of the DSP gas.

Finally estimating the typical k-value of the system via the inverse characteristic length scale, i.e. $k \sim 1/L_{\rm char}$ we can rewrite condition (19) and obtain

$$\frac{L_{\rm abs}}{L_{\rm char}} \leq \frac{\gamma}{|\Delta|}, \tag{20}$$

which is just condition (2) of the main text with the appproximation $\sin\theta \approx 1$.

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